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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

#12

AN RE APPLICATION OF

MANABU SUHARA ET AL

: EXAMINER: CANTELMO, G.

SERIAL NO: 10/089,109

FILED: MARCH 26, 2002

: GROUP ART UNIT: 1745

FOR: LITHIUM-COBALT COMPOSITE:

OXIDE, METHOD FOR PREPARING THE SAME, POSITIVE ELECTRODE FOR LITHIUM SECONDARY CELL AND LITHIUM SECONDARY CELL

APPEAL BRIEF

COMMISSIONER FOR PATENTS ALEXANDRIA, VA 22313

SIR:

This is an appeal of the Final Rejection dated July 29, 2003 of Claims 1, 2, 4 and 7-

14. A Notice of Appeal is submitted herewith.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is Seimi Chemical Co., Ltd. having an address at 3-2-10, Chigasaki, Chigasaki-City, Kanagawa 253-8585, Japan.

II. RELATED APPEALS AND INTERFERENCES

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Appellants, Appellants' legal representative and the assignee are aware of no appeals or interferences which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF THE CLAIMS

Claims 1, 2, 4 and 7-14 stand rejected and are herein appealed. Claims 5 and 6 stand objected to but drawn to allowable subject matter. Claim 3, the remaining claim in the application, stands withdrawn as being directed to a non-elected species.

IV. STATUS OF THE AMENDMENTS

No amendment under 37 CFR 1.116 has been filed.

V. SUMMARY OF THE INVENTION

As recited in Claim 1, the present invention is a hexagonal lithium-cobalt composite oxide for a lithium secondary cell, which is represented by the formula $\text{LiCo}_{1-x}M_xO_2$, wherein x is $0 \le x \le 0.02$ and M is at least one member selected from the group consisting of Ta, Ti, Nb, Zr and Hf, and which has a half-width of the diffraction peak for (110) face at $2 \theta = 66.5 \pm 1^\circ$, of from 0.070 to 0.180°, as measured by the X-ray diffraction using CuK_α as a ray source.

See the specification at page 4, lines 14-21.

VI. ISSUES

- (A) Whether Claims 1, 2 and 7-14 are anticipated under 35 U.S.C. § 102(b) by, or in the alternative, obvious under 35 U.S.C. § 103(a) over, JP-A-10-1316 (Aoki et al)?
- (B) Whether Claim 4 is unpatentable over <u>Aoki et al</u> in view of U.S. 5,709,969 (Yamahira)?
- (C) Whether Claims 1, 2, 7 and 10-14 are anticipated under 35 U.S.C. § 102(b) by, or in the alternative, obvious under 35 U.S.C. § 103(a) over, U.S. 5,147,738 (Toyoguchi)?
 - (D) Whether Claim 4 is unpatentable over Toyoguchi, in view of Yamahira?
- (E) Whether Claims 8 and 9 are unpatentable over <u>Toyoguchi</u> in view of U.S. 5,702,843 (<u>Mitate et al</u>)?
- (F) Whether Claims 1, 2, 7, 10 and 12-14 are provisionally unpatentable on grounds of obviousness-type double patenting over Claims 8-13 of copending Application No. 10/296,205 (copending application)?
- (G) Whether Claim 4 is provisionally unpatentable on grounds of obviousness-type double patenting over Claims 1 and 8-14 of the copending application in view of <u>Yamahira</u>?
- (H) Whether Claims 8, 9 and 11 are provisionally unpatentable on grounds of obviousness-type double patenting over Claims 1 and 8-14 of the copending application in view of Aoki et al?

VII. GROUPING OF THE CLAIMS

For issue (A), Claims 2 and 12 each stand or fall separately from independent Claim 1. Claim 14 stands or falls separately from independent Claim 13.

For issue (C), Claims 2 and 12 each stand or fall separately from Claim 1. Claim 14 stands or falls separately from Claim 13.

For issue (F), Claims 2 and 12 each stand or fall separately from Claim 1. Claim 14 stands or falls separately from Claim 13.

<u>VIII. ARGUMENT</u>

As a preface to discussion of the rejections over prior art, the present invention relates to a lithium-cobalt composite oxide for a lithium secondary cell, a process for its production, a positive electrode for a lithium secondary cell employing it, and a lithium secondary cell.

As described in the specification under "Background Art," beginning at page 1, line 11, lithium secondary cells employing a lithium-cobalt composite oxide (LiCoO₂) as a positive electrode active material, are known. Also known are such composite oxides wherein some amount of cobalt has been replaced with another element. Various processes for making such lithium-cobalt composite oxides are also known. One such process, as described in the specification at page 2, lines 6-12, and as disclosed in Aoki et al, supra, uses an active material LiCoO₂ which is obtained by dispersing in an aqueous lithium hydroxide solution, e.g., cobalt hydroxide or cobalt oxyhydroxide wherein the valence of cobalt is trivalent, followed by heat treatment, for the purpose of improving the cycle characteristics, etc., of a lithium secondary cell. Appellants further describe, at page 3, lines 10-17 of the specification, after discussing the prior art, that no product has been known which fully satisfies all of the requirements for cycle characteristics, the initial weight capacity density, the stability and the low temperature operation efficiency, and for a production method for efficient mass production.

As recited in Claim 1, the present invention is a hexagonal lithium-cobalt composite oxide for a lithium secondary cell, which is represented by the formula $\text{LiCo}_{1-x}\text{M}_x\text{O}_2$, wherein x is $0 \le x \le 0.02$ and M is at least one member selected from the group consisting of Ta, Ti,

Nb, Zr and Hf, and which has a half-width of the diffraction peak for (110) face at $2 \theta = 66.5 \pm 1^{\circ}$, of from 0.070 to 0.180°, as measured by the X-ray diffraction using CuK_a as a ray source.

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The Examiner has found that a process for producing the hexagonal lithium-cobalt composite oxide for a lithium secondary cell as defined in Claim 1 is patentable, as embodied in Claims 5 and 6.

The specification herein contains data showing that the value of "x" in LiCo_{1-x}M_xO₂ affects the half-width of the diffraction peak for (110) face at 2θ=66.5±1°. For example, Examples 15 and 16 each employ a value of x as 0.05, resulting in said half-width being higher than the presently-recited maximum of 0.180°, and also resulting in a capacity retention after 40 charge/discharge cycles, less than that of the corresponding example compared to, i.e., Examples 1 and 5, respectively. Thus, Example 1 had a value of x of 0.002, and a capacity retention after 40 charge/discharge cycles of 95.3%; that of Example 15 was 93.6%. Similarly, Example 5 had a value for x of 0.002, and a capacity retention of 95.8%; the capacity retention for Example 16 is 93.8%. In addition, the half-widths between Examples 1 and 15, and between Examples 5 and 16, respectively, differ.

In addition, each of Examples 12, 13 and 14 demonstrates how the change of a particular variable affects the results. Example 12 employs a cobalt oxide powder instead of cobalt oxyhydroxide as in Example 9. Example 13 employs a cobalt oxyhydroxide powder having a different average particle size and a different specific surface area compared to the cobalt oxyhydride powder used in Example 9. Example 14 employs a lower firing temperature than that used in Example 9. In all of these Examples 12-14, both the half-width and the capacity retention changed.

The above-discussed results could not have been predicted by the applied prior art.

In addition, it is understood that in preparation of lithium cobaltate, the properties of lithium cobaltate and characteristics of cells to be produced substantially depend on the types of the starting materials, properties of the starting materials, mixing method (wet system or dry system), and firing (sintering) temperature and atmosphere.

Issue (A)

Claims 1, 2 and 7-14 stand rejected under 35 U.S.C. § 102(b) as anticipated by, or in the alternative, under 35 U.S.C. § 103(a) as obvious over, <u>Aoki et al</u>. That rejection is untenable and should not be sustained.

In addition to the discussion above with regard to Aoki et al, Aoki et al disclose a lithium-cobalt composite oxide having the formula LiCo_{1-x}A_xO_y, wherein x is between 0 and 0.25, and y is between 1.875 and 2.25. A can be, *inter alia*, Ti, Nb, Zr, Hf, and Ta. However, Aoki et al do not exemplify any lithium-cobalt composite oxides within the terms of the present claims, and moreover, do not recognize the effect the value of "x" has, both on the half-width and the capacity retention. In addition, the above-discussed data in the specification shows that how the lithium-cobalt composite oxide is made affects its physical properties and thus, affects the make-up of the composite oxide itself.

The Examiner's rationale is that <u>Aoki et al</u> appears to disclose the claimed subject matter, and that the burden is on Appellants to show that the claimed subject matter is different from, and not suggested by, <u>Aoki et al</u>, relying on *In re Best*, 195 USPQ 430 (CCPA 1977).

In reply, Appellants do not dispute the indication in *Best* that the PTO can require a patent applicant to prove, where claimed subject matter is **reasonably** considered to be inherently or necessarily met by the prior art, that the claimed subject matter is not met by the

prior art (emphasis added). However, Appellants have demonstrated with the data of record that it is not reasonable to presume that Aoki et al meets, or otherwise suggests, the presently-claimed invention. Indeed, the CCPA in Best confirmed that indirect comparisons, based on established scientific principles, can validly be applied to distinguish a claimed chemical product from that disclosed in the prior art. Best, 195 USPQ at 432. While Appellants have not directly compared to Aoki et al, all the evidence of record, which shows the influence of many variables on the half-width of the diffraction peak, including the starting materials used, clearly suggest that Aoki et al's product does not meet the terms of the present claims.

Claim 2 is separately patentable, because <u>Aoki et al</u> neither disclose nor suggest the hexagonal lithium-cobalt composite oxide of Claim 1, wherein x is $0.0005 \le x \le 0.02$, and the half-width of the diffraction peak for (110) face is from 0.100 to 0.165° .

Claim 12 is separately patentable, because Aoki et al neither disclose nor suggest the hexagonal lithium-cobalt composite oxide for a lithium secondary cell according to Claim 1, which is obtained by a process which comprises dry blending a cobalt oxyhydroxide powder having an average particle size of from 1 to 20 µm and a specific surface area of from 2 to 200 m²/g, a lithium carbonate powder having an average particle size of from 1 to 50 µm and a specific surface area of from 0.1 to 10 m²/g, and a powder of an oxide of metal element M having an average particle size of at most 10 µm and a specific surface area of from 1 to 100 m²/gm, and firing the mixture at a temperature of from 850 to 1,000°C in an oxygencontaining atmosphere.

Similarly, Claim 14 is patentable, because <u>Aoki et al</u> neither disclose nor suggest the hexagonal lithium-cobalt composite oxide according to Claim 13, which is obtained by a process which comprises dry blending a cobalt oxyhydroxide powder having an average particle size of from 1 to 20 µm and a specific surface area of from 2 to 200 m²/g, a lithium

carbonate powder having an average particle size of from 1 to 50 μ m and a specific surface area of from 0.1 to 10 m²/g, and a powder of an oxide of metal element M having an average particle size of at most 10 μ m and a specific surface area of from 1 to 100 m²/gm, and firing the mixture at a temperature of from 850 to 1,000 °C in an oxygen-containing atmosphere.

As stated above, the Examiner has found that the recited process in Claims 12 and 14 is patentable.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Issue (B)

Claim 4 stands rejected under 35 U.S.C. § 103(a) as unpatentable over <u>Aoki et al</u> in view of <u>Yamahira</u>. That rejection is untenable and should not be sustained.

The Examiner relies on Yamahira for a disclosure of volumetric density, and specifically one having a range of from 2.0 to 4.3 g/ml (paragraph bridging columns 3 and 4). In Yamahira, a mixture of a lithium compound and a cobalt compound is compression molded, and then sintered to prepare a sintered compact pellet, and this pellet is as itself used as a positive electrode for e.g. a coin-shaped cell. In Yamahira, the volumetric density of the sintered compact pellet is from 2.0 to 4.3 g/ml. Accordingly, Yamahira is fundamentally different from the present invention in the method for producing the positive electrode material and the method for using the positive electrode material. In the present invention, a mixture is not pelletized and no sintered molded body is prepared. In the present invention, the press density of a sintered powder is defined. Nevertheless, even if the products of Aoki et al were to have the volumetric density of Yamahira, the result would still not be the

presently-claimed invention, because <u>Yamahira</u> does not remedy the above-discussed basic deficiencies of <u>Aoki et al.</u>

In the Final Office Action, the Examiner finds that Appellants have failed to "persuasively argue why it would not have been obvious to modify the primary reference in view of Yamahira." In reply, Aoki et al is not drawn to a sintered compact pellet which itself is used as a positive electrode. Thus, the volumeric density of Yamahira's sintered compact pellet has no relevance to the lithium-cobalt composite oxide of Aoki et al. Nor has the Examiner shown that packing press density is a result effective variable. Thus, In re Antonie applies (exceptions to rule that optimization of a result-effective variable is obvious, such as where the results of optimizing the variable are unexpectedly good or where the variable was not recognized to be result effective). Appellants are entitled to prevail under at least the second of the above exceptions.

Accordingly, it is respectfully requested that this rejection be REVERSED.

Issue (C)

Claims 1, 2, 7 and 10-14 stand rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as obvious over, <u>Toyoguchi</u>. That rejection is untenable and should not be sustained.

Toyoguchi discloses a cathode active material having the following formula:

$$Li_{X}Co_{(1-Y)}M_{Y}O_{2}$$
 (M=W, Mn, Ta, Ti, Nb),

which is prepared by mixing Li_2CO_3 , CoCO_3 and at least one oxide of W, Mn, Ta, Ti, Nb at predetermined ratio (Li atom = X mole, Co atom = 1-Y mole, M atom = Y mole), wherein $0.05 \le \text{Y} \le 0.35$, followed by heating the mixture at 900°C for 10 hours (column 1, lines 48-63 and column 2, lines 54-61). In each of Tables 1-5 therein, <u>Toyoguchi</u> exemplifies cathode

active materials of the above formula wherein, *inter alia*, X is 1, and Y is 0.02 (which examples are thus outside the scope of <u>Toyoguchi</u>'s invention). However, since <u>Toyoguchi</u> discloses preparing their cathode active material with starting materials different from the present starting materials, and in view of the above-discussed comparative data, it would not be expected that the cathode active material of <u>Toyoguchi</u> would be the same as, or have the same properties as, the presently-claimed lithium-cobalt composite oxide. Note also that when Y is within the terms of <u>Toyoguchi</u>'s invention, i.e., above 0.05, discharge capacity per unit weight tends to decrease.

It is clear that the Examiner's rationale for this rejection is the same as that for the rejection over Aoki et al, discussed under Issue (A), above. The points made therein in rebuttal apply herein as well.

In addition, the various claims that were argued separately under Issue (A) are separately patentable herein as well, for the same reasons as discussed under Issue (A), except that <u>Toyoguchi</u> applies herein rather than <u>Aoki et al</u>.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Issue (D)

Claim 4 stands rejected under 35 U.S.C. § 103(a) as unpatentable over <u>Toyoguchi</u> in view of <u>Yamahira</u>. That rejection is untenable and should not be sustained.

See the discussion under Issue (B) regarding the Examiner's reliance on <u>Yamahira</u>. Nevertheless, even if the products of <u>Toyoguchi</u> were to have the volumetric density of <u>Yamahira</u>, the result would still not be the presently-claimed invention, because <u>Yamahira</u> does not remedy the above-discussed basic deficiencies of <u>Toyoguchi</u>.

In the Final Office Action, it is clear that the Examiner's rationale for this rejection is the same as that for the rejection over <u>Aoki et al</u>, discussed under Issue (B), above. The points made therein in rebuttal apply herein as well, except that <u>Toyoguchi</u> applies herein rather than <u>Aoki et al</u>.

Accordingly, it is respectfully requested that this rejection be REVERSED.

Issue (E)

Claims 8 and 9 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Toyoguchi in view of Mitate et al. That rejection is untenable and should not be sustained.

The Examiner relies on <u>Mitate et al</u> for a disclosure of a current collector supporting an electrode. But even if a current collector were used in the cathode active material of <u>Toyoguchi</u>, <u>Mitate et al</u> would still not remedy the basic deficiencies of <u>Toyoguchi</u>, as discussed above.

Accordingly, it is respectfully requested that this rejection be REVERSED.

Issue (F)

Claims 1, 2, 7, 10 and 12-14 stand provisionally rejected on grounds of obviousness-type double patenting over Claims 8-13 of the copending application. That rejection is untenable and should not be sustained. Each of Claims 8-13 of the copending application depends on Claim 1 thereof, which reads as follows:

A lithium-transition metal composite oxide which comprises particles of a lithium composite oxide represented by the formula $\text{Li}_x M_{1.y} N_y O_2$, wherein $0.2 \le x \le 1.2$, $0 \le y \le 0.7$, M is a transition metal element, and N is a transition metal element other than M or an alkaline earth metal element, wherein in the distribution curve of the cumulative volume particle size of said lithium composite oxide, the inclination of the curve at a cumulative volume fraction of 20% is at most 9%/ μ m, the inclination of the curve at the cumulative

volume fraction of 80% is at least 3%/ μ m, and the average particle size is from 3 to 20 μ m.

Thus, the claimed subject matter in the copending application requires particular values of the inclination of the distribution curve of the cumulative volume particle size of the lithium composite oxide, one at a cumulative volume fraction of 20%, and one at a cumulative volume fraction of 80%. There are other differences as well. The subject matter of the present claims has no such requirement. The Examiner has no basis to conclude that the presently-claimed subject matter inherently or necessarily meets the two inclination limitations of the claims of the copending application.

Accordingly, it is respectfully requested that this rejection be REVERSED.

Issue (G)

Claim 4 stands provisionally rejected on grounds of obviousness-type double patenting over Claims 1 and 8-14 of the copending application in view of <u>Yamahira</u>. That rejection is untenable and should not be sustained.

Each of Claims 8-14 of the copending application depends on Claim 1 thereof, set forth under Issue (F), above.

Yamahira does not remedy any of the deficiencies discussed above under Issue (F).

In the Final Office Action, it is clear that the Examiner's rationale for this rejection is the same as that for the rejection over <u>Aoki et al</u>, discussed under Issue (B), above, and as that for the rejection over <u>Toyoguchi</u>, discussed under Issue (D), above. The points made therein in rebuttal apply herein as well, except that Claims 1 and 8-14 of the copending application apply herein rather than <u>Aoki et al</u> and <u>Toyoguchi</u>.

Accordingly, it is respectfully requested that this rejection be REVERSED.

Issue (H)

Claims 8, 9, and 11 stand provisionally rejected on grounds of obviousness-type double patenting over Claims 1 and 8-14 of the copending application in view of <u>Aoki et al</u>.

That rejection is untenable and should not be sustained.

Aoki et al does not remedy any of the deficiencies discussed above under Issue (F).

Accordingly, it is respectfully requested that this rejection be REVERSED.

IX. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending in the Final Office Action be REVERSED.

Respectfully submitted,

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APPENDIX

CLAIMS ON APPEAL

- 1. A hexagonal lithium-cobalt composite oxide for a lithium secondary cell, which is represented by the formula $\text{LiCo}_{1-x}\text{M}_x\text{O}_2$, wherein x is $0 \le x \le 0.02$ and M is at least one member selected from the group consisting of Ta, Ti, Nb, Zr and Hf, and which has a half-width of the diffraction peak for (110) face at $2\theta = 66.5 \pm 1^\circ$, of from 0.070 to 0.180°, as measured by the X-ray diffraction using CuK_α as a ray source.
- 2. The hexagonal lithium-cobalt composite oxide for a lithium secondary cell according to Claim 1, wherein x is $0.0005 \le x \le 0.02$, and the half-width of the diffraction peak for (110) face is from 0.100 to 0.165° .
- 4. The hexagonal lithium-cobalt composite oxide for a lithium secondary cell according to Claim 1, wherein the packing press density of the hexagonal lithium-cobalt composite oxide is from 2.90 to 3.35 g/cm³.
- 7. A positive electrode for a lithium secondary cell, which contains the hexagonal lithium-cobalt composite oxide for a lithium secondary cell as defined in Claim 1, as an active material.
- 8. The positive electrode for a lithium secondary cell according to Claim 7, having a mixture comprising the active material, an electrically conductive material and a binder, supported on a current collector.
- 9. The positive electrode for a lithium secondary cell according to Claim 8, wherein the current collector is aluminum or stainless steel.
- 10. A lithium secondary cell employing a positive electrode which contains the hexagonal lithium-cobalt composite oxide for a lithium secondary cell as defined in Claim 1, as an active material.

- 11. The lithium secondary cell according to Claim 10, wherein a cyclic or chain carbonic ester is used as a solvent for the electrolyte.
- 12. The hexagonal lithium-cobalt composite oxide for a lithium secondary cell according to Claim 1, which is obtained by a process which comprises dry blending a cobalt oxyhydroxide powder having an average particle size of from 1 to 20 μm and a specific surface area of from 2 to 200 m²/g, a lithium carbonate powder having an average particle size of from 1 to 50 μm and a specific surface area of from 0.1 to 10 m²/g, and a powder of an oxide of metal element M having an average particle size of at most 10 μm and a specific surface area of from 1 to 100 m²/gm, and firing the mixture at a temperature of from 850 to 1,000°C in an oxygen-containing atmosphere.
- 13. A hexagonal lithium-cobalt composite oxide for a lithium secondary cell, which is represented by the formula $\text{LiCo}_{1-x}\text{M}_x\text{O}_2$, wherein x is $0 < x \le 0.02$ and M is at least one member selected from the group consisting of Ta, Ti, Nb, Zr and Hf, and which has a half-width of the diffraction peak for (110) face at $2 \theta = 66.5 \pm 1^\circ$, of from 0.070 to 0.180°, as measured by the X-ray diffraction using CuK_α as a ray source.
- 14. The hexagonal lithium-cobalt composite oxide for a lithium secondary cell according to Claim 13, which is obtained by a process which comprises dry blending a cobalt oxyhydroxide powder having an average particle size of from 1 to 20 μm and a specific surface area of from 2 to 200 m²/g, a lithium carbonate powder having an average particle size of from 1 to 50 μm and a specific surface area of from 0.1 to 10 m²/g, and a powder of an oxide of metal element M having an average particle size of at most 10 μm and a specific surface area of from 1 to 100 m²/gm, and firing the mixture at a temperature of from 850 to 1,000°C in an oxygen-containing atmosphere.